From Earth s Building Blocks to Metallic Planetary Cores

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Summary

This thesis combines the fields of experimental petrology and non-traditional stable isotope geochemistry to study silicon (Si) isotope fractionation between metal and silicate phases over a wide range of pressure and temperature conditions. The aim of this work was to develop a tool for tracing planetary accretion and metal core formation processes on the basis of experimentally determined Si isotope fractionation behaviour in metal-silicate systems.

The main conclusions of this thesis are contained in chapters five, six and seven. A rather untraditional experimental approach was taken in chapter five, in which Si isotope fractionation between metal and silicate was studied on samples obtained from a metal-alloy producing industrial-scale blast furnace at Tata Steel IJmuiden, the Netherlands. In this chapter the extent and mechanisms of Si isotopic fractionation at conditions relevant to metal segregation in small planetary bodies were assessed. A model was developed for metal-silicate Si isotope fractionation in blast furnaces, which can explain both the sense and magnitude of fractionation if the presence of an SiO-bearing vapour phase is taken into account. The results of this study were applied to models of aubrite meteorite formation through high-temperature differentiation of an enstatite chondrite parent body. Our data suggest a far larger degree of re-homogenization during differentiation than previously thought on the basis of metal-silicate Si isotopic fractionation measured in natural meteorites that re-equilibrated at low temperature.

In chapter six, the kinetic isotope fractionation behaviour of Si was investigated in the one-atmosphere gas-mixing furnace set up at VU University Amsterdam (as described in chapter two). In two experimental series at 1400^{o}C and 1550^{o}C the oxidation of elemental Si to silicate is simulated by exposing starting material consisting of a high Si-bearing, highly reduced metal phase and fayalite to less reducing conditions. This approach was taken to improve the understanding of micro and global scale metal-silicate equilibration processes at pressure-temperature conditions relevant to Si-bearing metal phases entering a shallow magma ocean. The experiments show that kinetic Si isotopic fractionation is as large as, but opposite in sign to, equilibrium Si isotope fractionation in metal-silicate systems. These results demonstrate that Si diffusion can cause significant kinetic Si isotope fractionation. Results of this study suggest that Si isotope measurements in meteorite metal phases can be used to assess the role of kinetic versus equilibrium processes during the initial formation of meteorites at high temperatures and their subsequent alteration at lower temperature.

In chapter seven, the Si isotope fractionation between metal and silicate was studied in two experimental high-pressure and high-temperature (*HPT*) series. One series used Si-bearing Fe metal in the starting material; the other series used Al-bearing Fe. The experiments were performed in the multi-anvil devices of the *HPT* laboratories of Bayerisches Geoinstitut (BGI) in Bayreuth, Germany. Our data extend the previously published *HPT* data set (1-7 GPa) to higher-pressure conditions (9 GPa), that enable a negative pressure dependence on metal-silicate Si isotope fractionation to be identified for the first time. Incorporating this pressure effect into terrestrial core formation models further complicates estimates of the Si content of the core based on the mismatch in Si isotopic composition between the Bulk Silicate Earth (BSE) and primitive meteorites. The

maximum Si content of the core based on geophysical measurements and mineral physics data is approximately 10 weight per cent. This can only be reconciled with our isotope fractionation measurements if the difference between the Si isotopic composition of BSE and the starting materials of the Bulk Earth is smaller than virtually all current estimates, or if the pressure of equilibration between metal and silicate during core formation was much lower than currently thought.

The work presented in chapters five through seven indicates that additional experimental data will be required to fully characterize the fractionation behaviour of Si isotopes in *HPT* systems. The results presented in this thesis suggest that temperature, pressure, chemical composition, physical state, and time can all influence Si isotope fractionation between metal and silicate significantly. Results show the importance of accurate understanding of isotope fractionation mechanisms at *HPT* conditions. As detailed below, the findings of this thesis point to several research directions that should be pursued to further improve Si isotopic constraints on accretion and core formation processes.